Charge at Ferroelectric Interfaces

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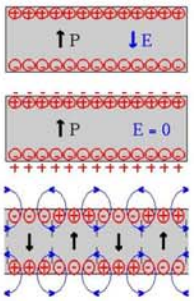
Motivation:

- Compensation of the depolarizing field by charge at interfaces is critical for functioning of standard ferroelectric devices (e.g., nonvolatile memories)
- Compensation is also potential mechanism for coupling ferroelectric behavior with the chemical environment
- Standard models of interfacial charge do not explain recently observed behavior in thin films



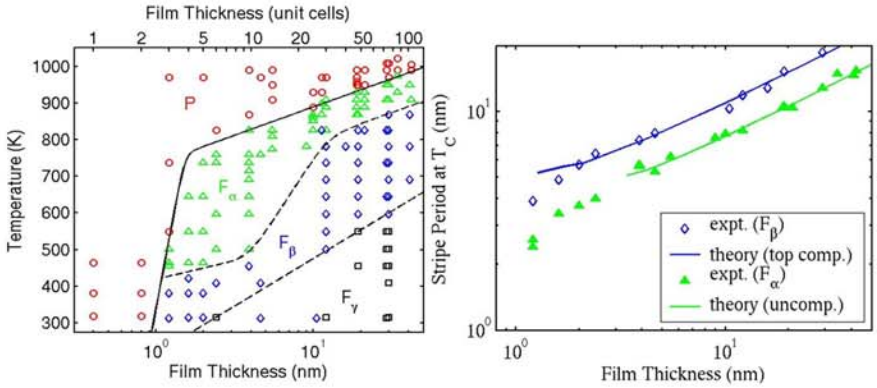
Depolarizing field *E* opposes *P*

E compensated by free charge

E compensated by stripe domains

- Challenge: understand the nature of the compensating charge
- Our approach:
  - Use *in situ* synchrotron x-ray scattering to observe ferroelectric behavior as a function of thickness, temperature, electrical boundary conditions, and environment
  - Compare x-ray results to both mean field and *ab initio* theory

Major Accomplishment: Phase Diagram of Equilibrium 180° Stripe Domains

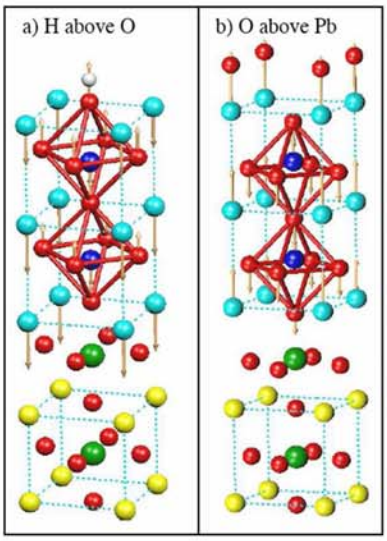


Impact:

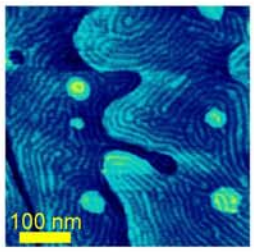
- Observe two 180° stripe domain phases (F_α, F_β) and one monodomain phase (F_γ)
- Equilibrium stripe structure in good agreement with Landau theory (Stephenson & Elder, to be published in *J. Appl. Phys.*)
- Results suggest intrinsic surface effect may *enhance* ferroelectric behavior at nanoscale
- Transitions from F_α to F_β and F_β to F_γ reflect sequential compensation of upper and lower interfaces - possible method for controlling ferroelectricity with interfacial chemistry and vice versa

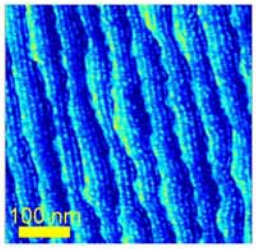
Future Directions:

- Results from preliminary experiments and *ab initio* theory confirm adsorbed ions can compensate upper interface

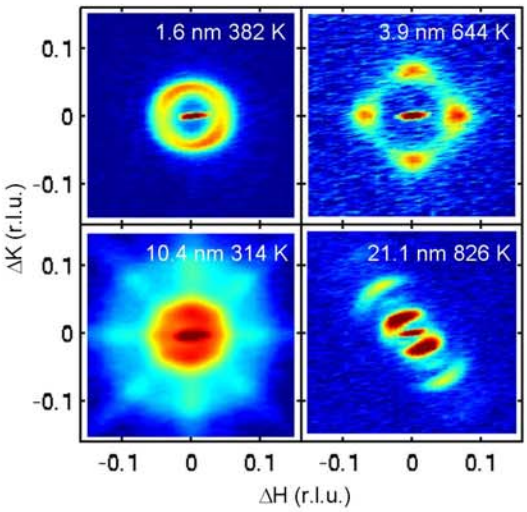


- Implications
  - We can switch polarization by changing chemical environment
  - We can change adsorption behavior by poling ferroelectric
  - Adsorbates self-assemble on 180° stripe domains





- In-plane reciprocal space maps indicate strong dependence of stripe domain morphology on film thickness, temperature, cooling rate and crystal miscut



- 180° stripe domains interact with steps on $PbTiO_3$ surface → path to controlling morphology and templated self-assembly

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